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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The primary objective of this research program was to assess and develop the use of high-precision ultrasonic interferometry as a means of evaluating the internal residual stresses in polycrystalline materials. The method which was pursued involved the comparison of the elastic properties of the polycrystalline solid with those inferred for the stress-free aggregate from single-crystal data used in conjunction with an appropriate theoretical averaging scheme. Another approach, based on residual stress-induced transverse isotropy,		

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also was evaluated. A primary problem with all acoustic approaches is associated with effects arising from microstructural characteristics. In the present study, accurate microstructure characterization and theoretical models were utilized to account for these effects. Residual stress evaluation by the photoelastic method was employed also as an independent means of comparison. Work in this study was limited to  $\text{MgAl}_2\text{O}_4$  spinel, which was available in high-quality single-crystal specimens and polycrystalline blanks.

Extensive sets of elastic property data were obtained as functions of both temperature and pressure for numerous specimens of single-crystal and polycrystalline spinel. Our single-crystal second-order elastic constant results for stoichiometric spinel compared favorably with those from previous studies. The higher-order property results, particularly with respect to temperature, are new and provide useful insight relevant to the high P,T equation of state. Aggregate "average" elastic properties, evaluated from the single-crystal data using the Hashin-Shtrikman bounding approach, provided the basis for the residual stress assessment in the polycrystalline specimens. The residual stress levels in the latter were evaluated by photoelastic methods to be less than about 25 MPa. Ultrasonic interferometry acoustic velocities suggested residual stresses in the polycrystalline spinel specimens which were an order of magnitude higher. However, microstructure characterization combined with theoretical modelling indicate that the apparent high values of residual stress suggested by the ultrasonic data are actually primarily the result of grain-pore texture. Therefore, in single-phase polycrystalline aggregates of cubic crystallites, where the actual residual stress levels are likely to be very low, acoustic methods are difficult to apply because of competing textural effects.

## Residual Stresses and Elastic Properties in Polycrystalline Materials

## Final Report

E.K. Graham and R.C. Bradt

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FINAL REPORT: RESIDUAL STRESSES  
AND ELASTIC PROPERTIES IN POLYCRYSTALLINE MATERIALS

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Statement of the Problem

The elastic properties of polycrystalline materials are of fundamental significance to materials science, as well as other physical sciences, including geology and the engineering disciplines. The elastic behavior of a dense polycrystalline body depends on the composition, porosity, grain size and texture or orientation, as well as the nature of the densification procedure. The latter is responsible, to a large extent, for the residual internal stresses which characterize many hot-pressed polycrystalline materials. Of course, thermal expansion anisotropy in non-cubic systems is also of consequence. The significance of the foregoing polycrystalline characteristics are widely recognized in a qualitative sense; However, a rigorous, quantitative understanding of the physical effects of such features has not been achieved. This is especially true in the case of residual stresses.

Residual stresses in materials can play many roles--they may be beneficial, detrimental, or of an indeterminate character. Although much of the activity concerned with quantitative residual stress evaluation has been

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focused on metallic materials, a considerable amount of recent effort has been directed toward ceramics, especially glasses. Both ceramics and glasses with superior strength have been engineered by considering residual stress development. In particular, residual compressive stresses can be highly beneficial in inhibiting failure, as has been amply demonstrated on a commercial basis for thermally and chemically tempered glass artifacts. Residual stresses are not only important with regard to mechanical design considerations, they are also important with regard to optical, electric, and magnetic properties. Magnetostriction and piezoelectric effects are examples of the coupling of stresses to the latter two groups of properties. The use of stress birefringence for stress measurement and the coupling of stress and light in acousto-optic devices are examples of the former. In a manner analogous to the failure criterion equation, the application of stress from an external source will add to those internal residual stresses that are present, so it is of consequence to be able to assess the state of residual stress for practically every critical property application, whether it be mechanical, magnetic, or optical.

Considering the importance of residual stresses in ceramic and metallic material mechanical and physical properties, as well as in materials engineering, it is necessary to develop techniques to measure and evaluate internal and near-surface stress levels, preferably in a non-destructive manner. In this regard, a number of methods have been proposed and used. The most common technique which has been used to evaluate "micro"-stresses (short-range stress concentrations) is X-ray diffraction. Longer range (or "macro") stresses also can be assessed by X-ray diffraction by comparing d-spacings to those of the relaxed state. Unfortunately, because of the limited penetration of the radiation, X-ray diffraction tends to be most

useful to assess near-surface stresses only. Several optical methods also have been devised for measuring residual stresses, including laser-light scattering, laser speckling, and photoelasticity. The latter involves the dependence of the index of refraction on stress. It is very sensitive and provides an effective way of measuring very small residual stresses. However, a severely limiting aspect of the method is the need for transparent specimens. Another class of methods for residual stress evaluation utilizes material elastic properties as defined by ultrasonic waves. These techniques are more effective for assessing internal macrostresses. The very high precision of ultrasonic interferometry suggests the potential for resolving very small levels of stress. However, unfortunately, the high sensitivity of the acoustic methods is offset by the strong dependence of elastic properties (and wave velocities) on microstructural characteristics; in particular, porosity, pore and grain shape, pore and grain orientation, and even moisture content. In order to evaluate residual stresses in polycrystalline ceramic materials using ultrasonic methods, the effects due to these microstructure characteristics must first be assessed and removed. In addition to X-ray diffraction, optical, and acoustic methods, attempts to measure residual stresses also have been made using neutron diffraction, impurity band broadening, and magnetic methods. However, success with these methods has been very limited as yet.

The general purpose of this project has been to assess and evaluate the potential value of acoustic methods in the characterization of residual stresses in polycrystalline materials. In particular, attention was focused on the effects of residual stresses on elastic properties as defined by the measurement of ultrasonic wave velocities by precise ultrasonic interferometry. An effort was made to discriminate between the residual

stress-related effects and those resulting from microstructure characteristics. The present study efforts were limited to hot-pressed polycrystalline magnesium aluminate spinel ( $\text{MgAl}_2\text{O}_4$ ). Excellent specimens of both single-crystal and polycrystalline spinel are readily available from commercial sources. Moreover, spinel is transparent, easy to prepare for the necessary measurements, and has applications as a window and engineering material.

### Summary of the Most Important Results

#### Approach and Procedures

In order to evaluate levels of internal residual stress in a polycrystalline aggregate, it is necessary to establish a reference model in terms of an ideal material. The latter would be characterized by perfect homogeneity and isotropy (relative to the characteristic length of the experiment), zero residual stress, and zero porosity. The elastic moduli for the ideal model material must be evaluated, as well as their dependence on stress (pressure) and temperature. In the present study, we chose to meet the foregoing requirement by making the appropriate measurements on high-quality single-crystal  $\text{MgAl}_2\text{O}_4$  spinel, and then using our single-crystal elastic constant results in a suitable theoretical averaging scheme to define the ideal polycrystal properties. This approach avoids the microstructure problems inherent in real polycrystalline specimens.

For the foregoing reason, one of the primary aspects of the project has involved the accurate and precise measurement of the elastic constants (stiffnesses  $C_{ij}$  and compliances  $S_{ij}$ ) of single-crystal  $\text{MgAl}_2\text{O}_4$  at high temperature and pressure. These data provide the basis for the evaluation of the pressure derivatives of the individual elastic constants, as well as their temperature dependence [e.g.  $(\partial C_{ij}/\partial T)_P$ ,  $(\partial C_{ij}/\partial P)_T$ ,  $(\partial^2 C_{ij}/\partial P \partial T)$ ]. In turn,



the foregoing parameters provide the necessary input information for determining the elastic behavior of the ideal polycrystalline aggregate. In addition, they may be used in a suitable equation of state in order to specify accurately volume or density within a particular P, T regime.

The single-crystal specimens used in the present study were obtained from a large boule of Czochralski-grown stoichiometric  $\text{MgAl}_2\text{O}_4$  ( $\langle 100 \rangle$  growth axis) which was acquired from the Crystal Products Division of the Union Carbide Corporation. Each specimen was prepared in the configuration of a rectangular parallelepiped with faces oriented perpendicular (by means of Laue back-reflection X-ray diffraction) to the  $[100]$  and  $[110]$  crystallographic directions. The ultrasonic wave velocities were measured as functions of pressure and temperature using the precise Pulse Superposition (PS) method<sup>1,2</sup> of acoustic interferometry. A variation of Cook's method<sup>3,4</sup> was used to evaluate the elastic moduli parameters from the basic acoustic data. The pressure and temperature measurements were carried out within an internally heated Argon gas pressure vessel<sup>5,6</sup>. Data were obtained up to 1 GPa (10 kbar) with pressure defined by a calibrated manganin coil and digital monitoring system. Temperature measurements were carried out over the range 15 to 60°C using chromel-alumel thermocouples.

The single-crystal elastic moduli  $C_{ij}(P,T)$  were used to calculate the ideal polycrystal elastic properties using the VRH<sup>7-9</sup> and Hashin-Shtrikman<sup>10-13</sup> averaging techniques. The latter have been demonstrated<sup>14</sup> to yield the tightest possible bounds on the polycrystalline bulk modulus, given only the moduli and volume fractions of the constituent phases. Although a rigorous proof has not been given for the shear (rigidity) modulus<sup>13</sup>, we assume that a similar situation holds. In order to improve upon the Hashin-Shtrikman bounds, more information is required concerning the grain shape and size distribution.

For cubic materials, the upper and lower bounds on the bulk modulus are equivalent; moreover, the shear modulus bounds differ by less than 4%. Therefore, we regard the Hashin-Shtrikman bound averages as an accurate representation of the polycrystalline moduli.

The polycrystalline specimens used in the project were hot-pressed magnesium aluminate spinel obtained from the Coor's Porcelain Company. For proprietary reasons, no information was provided concerning the specific procedure and conditions of fabrication of the material. The specimens were cut and prepared from a single large disk-shaped blank 11.5 cm in diameter and 2.5 cm in thickness. A smaller transparent hot-pressed specimen was provided also by Coors. Some of the optical and microstructure studies were carried out on this specimen. In order to provide sufficient material for the large variety of physical property measurements required by this project, and to check for lateral variations, a number of specimens were prepared from the large blank.

Because of the necessity for assessing the effects on the polycrystalline elastic moduli due to microstructural properties, the hot-pressed blank specimens were characterized carefully using a variety of techniques. In particular, composition, pore volume (porosity) and shape distribution, grain size distribution and orientation, and gradients in these variables, were considered relevant aspects of the problem. In this regard, the following characterization procedures were carried out:

- 1) X-ray measurements were performed using an automated Rigaku diffractometer. These analyses were carried out to establish the nature of the phases present, evaluate the lattice parameters, and assess the extent and direction of preferred grain orientation.

- 2) A sample from the hot-pressed blank was submitted to the Mineral

Constitution Laboratory of The Pennsylvania State University for a quantitative chemical analysis.

3) Optical microscopy was used to evaluate grain size and structure on both hot-pressed blank and transparent polycrystal specimens. Several reflected light micrographs were taken to observe the characteristics of the center portion and edge regions of the former.

4) Scanning Electron Microscopy (SEM) also was performed on the specimens prepared for the optical observations in order to assess smaller scale microstructure.

5) Measurements of density are required in order to calculate the elastic moduli, as well as to evaluate porosity. The usual Archimedes' method using distilled water was employed for most of the density measurements.

6) Mercury intrusion porosimetry also was used to evaluate porosity and the pore size distribution. Scans were carried out from ambient pressure to 0.4 GPa on a series of large fragments and coarse powder specimens from the hot-pressed blank.

Following complete characterization of the microstructure of the hot-pressed blank specimens using the foregoing techniques, their effects on the elastic moduli were evaluated using several theoretical approaches. Of primary concern are the effects associated with porosity and preferred orientation of mineral grains and/or pore structure. In order to interpret the ultrasonic wave velocities, we employed nonlinear elasticity theory for transverse isotropy<sup>15,16</sup>. Models were tested which ascribed the observed experimental results to uniaxial residual stress,<sup>16,17</sup> preferred orientation of mineral grains,<sup>18,19</sup> pore volume and geometry,<sup>20,22</sup> preferred orientation of pore structure,<sup>23</sup> and compositional inhomogeneity. The preceding

microstructural data on the polycrystalline spinel material provided quantitative constraints in the evaluation of the various models.

The ultrasonic wave velocities (elastic properties) of the  $\text{MgAl}_2\text{O}_4$  polycrystalline specimens also were measured by PS acoustic interferometry. The pressure and temperature range and experimental method was similar to that of the single-crystal specimens, as described previously. As indicated in a preceding section, the primary purpose of this investigation was to assess the value of acoustic methods in the detection and evaluation of residual stresses in polycrystalline aggregates. However, because of the transparent and isotropic nature of the spinel samples, they were also suitable for a determination of residual stresses using the photoelastic effect<sup>24</sup>. Polarized light phase shifts due to stress-induced index of refraction variations were measured using the sensitive Relative Retardation technique. These data provided an independent evaluation of the polycrystalline spinel residual stress levels for comparison with the ultrasonic results. Because both methods reflect large volume macrostresses, this constitutes a very significant means of comparison.

#### Single-Crystal Results

The elastic properties of stoichiometric single-crystal spinel measured in this project constitute the subject matter of the M.S. thesis of Eric G. Hilbert. A copy already has been forwarded to the ARO. In addition, some further results on the higher-order elastic constants will be reported in the forthcoming Ph.D. thesis of Mr. Hilbert. The single-crystal results also are the subject of a forthcoming paper, which will be submitted for publication in the near future. A summary of the major results of the single-crystal spinel work follows.

The results for the adiabatic second-order elastic stiffness constants are  $C_{11} = 282.11 \pm 0.15$ ,  $C_{12} = 155.97 \pm 0.15$ , and  $C_{44} = 154.40 \pm 0.04$  GPa. The bulk modulus was determined to be  $198.02 \pm 0.15$  GPa. These results generally agree with those of previous studies. However, they represent a considerable improvement in accuracy over the values reported in the standard reference on elastic constants by Simmons and Wang<sup>25</sup>, which are due to Lewis<sup>26</sup>.

Measurements were also conducted at elevated hydrostatic pressures up to approximately 1.0 GPa. Ultrasonic travel times were best described by linear functions of pressure. The isothermal pressure derivatives of the second-order adiabatic stiffness coefficients were found to be  $4.474 \pm 0.029$ ,  $4.166 \pm 0.029$ , and  $0.8435 \pm 0.0023$  for the 11, 12, and 44 components, respectively. The isothermal pressure derivative of the bulk modulus was determined to be  $4.269 \pm 0.029$ . These results disagree with those of O'Connell and Graham<sup>27</sup>, whose generally lower values are probably due to complications typical of ultrasonic buffer rod techniques. The results disagree even more strikingly with those of Chang and Barsch<sup>3</sup>; in addition to reporting systematically higher pressure derivatives, they measured anomalously large second pressure derivatives, e.g.,  $-4.8 \text{ GPa}^{-1}$  for the bulk modulus. Such values were resolvable in this study, but were not observed. As determined by this study, the second pressure derivatives, if non-zero, were smaller in magnitude than  $-1.0 \text{ GPa}^{-1}$ ; this conclusion is qualitatively consistent with central-force lattice dynamical models. It implies that shear softening is not a dominant process in the high-pressure elastic behavior of spinel.

As a function of temperature, the ultrasonic travel times were linear. The isobaric temperature derivatives of the second-order adiabatic stiffness coefficients were measured to be  $-0.02549 \pm 0.00030$ ,  $-0.00944 \pm 0.00030$ , and  $-0.01011 \pm 0.00009 \text{ GPa/K}$  for the 11, 12, and 44 components, respectively. The

isobaric temperature derivative of the bulk modulus was determined to be  $-0.01479 \pm 0.00030$  GPa/K. These results are in partial agreement with the results of Chang and Barsch<sup>3</sup>; they are in complete agreement with the results of Liu et al.<sup>28</sup>, who used a somewhat less precise Raman-Nath scattering technique. The results of O'Connell and Graham<sup>27</sup> are systematically higher in magnitude.

Measurement of the pressure dependence of the temperature derivatives were not as conclusive as desired, the mixed second derivatives of the second-order adiabatic stiffness coefficients only being constrained as follows:  $-2.9$  to  $+6.5$ ,  $-12.1$  to  $+6.8$ , and  $-8.9$  to  $+0.53 \times 10^{-6} \text{ K}^{-1}$  for the 11, 12, and 44 components, respectively. The bulk modulus was constrained to the range  $-5.9$  to  $+6.7 \times 10^{-6} \text{ K}^{-1}$ . These values are smaller than the mixed second derivatives reported for other earth-forming materials by two orders of magnitude<sup>4</sup>.

Voigt, Reuss, and Hill (VRH) averages were computed for the bulk and shear moduli of polycrystalline spinel. The Hill average shear modulus was  $107.82 \pm 0.12$  GPa. The Hashin-Shtrikman bounds were also evaluated. These values for the shear modulus were  $109.95 \pm 0.17$  and  $106.34 \pm 0.27$  GPa for the upper and lower bound, respectively. The bulk modulus was  $198.02 \pm 0.15$  GPa; due to cubic symmetry, the same value applies for all averaging methods and both bounds.

#### Polycrystalline Elastic Results

The elastic property results for the polycrystalline spinel specimens, as well as the assessment of the residual stress properties of these materials, are the subject matter of the Ph.D. thesis of Blain Olbert, which is presently in the final stages of revision. A paper for publication is also planned.

Ultrasonic wave velocities were measured on five different specimens (rectangular parallelepipeds approximately 1/2 cm on edge) taken from the

center region of the polycrystalline spinel blank provided by Coor's Porcelain. Measurements also were made on a large rectangular slab cut from the blank across a diameter section. The five smaller specimens were cut with two of the faces parallel to the hot-pressed surface of the blank, and the remaining two pair of faces normal to the blank's hot-pressed surface. Five different wave velocities were measured in the cubes; two longitudinal and three shear modes. The longitudinal velocities,  $v_1$  and  $v_2$ , were evaluated for propagation directions parallel ( $v_1$ ) and perpendicular ( $v_2$ ) to the hot-pressing direction; the shear velocities ( $v_3$ ,  $v_4$ , and  $v_5$ ) were determined for propagation parallel to the hot-pressing direction ( $v_3$ , arbitrary polarization direction) and perpendicular to the hot-pressing direction ( $v_4$ , polarization parallel to the hot-pressing direction). Only those velocities corresponding to  $v_2$ ,  $v_4$ , and  $v_5$  were measured in the slab specimen. The small specimen compressional wave velocities ( $v_1$  and  $v_2$ ) differed from one another, as did the shear wave velocities ( $v_3$ ,  $v_4$ , and  $v_5$ ). However, the velocities  $v_2$ ,  $v_4$ , and  $v_5$  were nearly equivalent for the two propagation directions normal to the hot-pressing direction of the blank. For this reason the specimens were regarded for convenience as transversely isotropic.

The compressional and shear wave velocities varied in a continuous and systematic manner through the polycrystalline blank. They were generally greater in regions near the hot-pressed surfaces of the original blank, and smaller toward the center of the blank. The variation in acoustic velocities with respect to distance from the top hot-pressed surface of the blank (denoted by the variable  $Z$ ) was nearly parabolic in shape and symmetric about the center point of the blank. The average velocities in the small specimens varied between the limits:  $\bar{v}_1$  (at  $Z = 0$ ) = 9.790 km/sec,  $\bar{v}_1$  ( $Z = 1.2$  cm) = 9.745 km/sec;  $\bar{v}_2$  ( $Z = 0$ ) = 9.795 km/sec,  $\bar{v}_2$  ( $Z = 1.2$  cm) = 9.770 km/sec;

$\bar{v}_3 (Z = 0) = 5.555 \text{ km/sec}$ ,  $\bar{v}_3 (Z = 1.2 \text{ cm}) = 5.545 \text{ km/sec}$ ;  $\bar{v}_4 (Z = 0) = 5.536 \text{ km/sec}$ ,  $\bar{v}_4 (Z = 1.2 \text{ cm}) = 5.530 \text{ km/sec}$ ;  $\bar{v}_5 (Z = 0) = 5.555 \text{ km/sec}$ ,  $\bar{v}_5 (Z = 1.2 \text{ cm}) = 5.547 \text{ km/sec}$ . Similar velocities were measured in the slab. The very small, but definitive discrepancies, attest to the high accuracy and precision of the ultrasonic interferometry technique.

In contrast to the velocities, the lattice parameters ( $a_0$ ), determined as a function of  $Z$ , failed to change significantly. The value  $a_0$  ranged from only 0.80771 nm to 0.80762 nm, with a mean value of  $0.80765 \pm 0.00008 \text{ nm}$  (the uncertainty represents the 95% Confidence Interval about the mean). However, the bulk densities of the specimens taken from the polycrystalline blank changed with  $Z$  in a manner very similar to that of the velocities. The density variation was approximately symmetric about the center of the billet, with  $\rho(Z = 0) = 3.5744 \text{ g/cm}^3$  and  $\rho(Z = 1.2 \text{ cm}) = 3.5714 \text{ g}$ . Therefore, it appears that the systematic variation in the ultrasonic wave velocities correlates with, and is related to, a systematically varying porosity in the polycrystalline blank. Further support for this interpretation is provided by observations by Scanning Electron Microscopy. The predominant porosity was isolated on grain boundaries, with the pores appearing to be wedge-shaped and extending over an entire grain length. The mean grain size was about 36 microns, and the average pore width range was approximately 0.9 - 1.6 microns. Evaluation of the level of porosity from the differences between measured bulk and theoretical densities indicated an average value of only about 1/4%. As a result, the average polycrystalline wave velocities (9.775 and 5.545 km/sec for compressional and shear waves, respectively) are very similar to the HS average values (9.778 and 5.497 km/sec) from our single-crystal data, as well as from other studies<sup>5, 26, 28</sup>. The slight



discrepancies most likely arise as a result of the combination of porosity and/or stoichiometry differences.

#### Evaluation of Residual Stresses

In addition to the ultrasonic wave velocities, the piezo-optical coefficients of single-crystal and polycrystalline spinel were measured in order to evaluate residual stresses using the photoelastic effect. The single-crystal piezo-optical coefficients were found to be  $q_{11} = (2.693 \pm 0.024) \times 10^{-13} \text{ m}^2/\text{N}$ ,  $q_{12} = -(3.3103 \pm 0.026) \times 10^{-13} \text{ m}^2/\text{N}$ , and  $q_{44} = (5.908 \pm 0.112) \times 10^{-13} \text{ m}^2/\text{N}$ . The aggregate Voigt-Reuss-Hill values calculated from the foregoing single-crystal piezo-optical coefficients are given by  $\bar{q}_{11}^{\text{VRH}} = 2.795 \times 10^{-13} \text{ m}^2/\text{N}$  and  $\bar{q}_{12}^{\text{VRH}} = -3.2265 \times 10^{-13} \text{ m}^2/\text{N}$ , yielding,  $(\bar{q}_{11} - \bar{q}_{12})^{\text{VRH}} = 6.021 \times 10^{-13} \text{ m}^2/\text{N}$ . The mean piezo-optical coefficient difference for transparent, polycrystalline spinel was measured to be  $(\bar{q}_{11} - \bar{q}_{12}) = 6.080 \pm 0.089) \times 10^{-13} \text{ m}^2/\text{N}$ . Using the experimentally determined value of  $(\bar{q}_{11} - \bar{q}_{12})$ , residual stresses were evaluated for the center region of the slab sample. The residual stress distribution was symmetric about the center of the billet. The stresses ( $\sigma$ ) determined in the same region from which the acoustic wave velocities were measured indicated a change from -22 MPa at the hot-pressed surface of the polycrystalline blank to zero at the center. Thus, the photoelastic effect residual stress evaluation, which is accurate and not dependent on microstructural variations, indicates maximum values of magnitude < 25 MPa for the polycrystalline spinel sample.

Assuming that the velocity anisotropy (transverse isotropy) is the result of an internal residual stress of uniaxial character, one can estimate the level of the stress by

$$\sigma = \rho_0 (v_3^2 - v_4^2),$$

where  $\rho_0$  is the ambient density, and  $v_3$  and  $v_4$  are shear wave velocities for propagation parallel (arbitrary polarization) and perpendicular (polarized parallel to the stress direction) to the stress direction. Using the velocities measured on the specimens from the polycrystalline blank, yields results of 710 and 600 MPa for the surface and center regions of the blank. Clearly, these results are over an order of magnitude higher than those provided by the photoelastic effect. Moreover, they are most likely much higher than the load pressures used in the hot-pressing procedure. Thermal effects can not be a factor because spinel is cubic and therefore isotropic in its thermal expansion properties. Thus, it appears that the velocity results do not directly reflect the internal residual stresses in the spinel polycrystalline material. Rather, we conclude that the observed velocity anisotropy is primarily a textural effect. In the present case, in fact, a theoretical model which incorporated a slight preferred grain orientation together with a pore structure of random spheroidal voids, was able to explain all the velocity data reasonably well.

Therefore, our results indicate that in the case of polycrystalline aggregates of cubic grains, ultrasonic interferometry does not provide an accurate assessment of internal residual stress. This is not necessarily because of insufficient precision. Rather, the root of the problem lies in the difficulty in discriminating the lower levels of residual stress from the dominant effects of texture on the acoustic waves. Only in cases where the latter are demonstrably negligible, or may be accounted for by accurate microstructure characterization together with theoretical modelling, can the residual stress levels be evaluated with confidence. For cubic grain polycrystalline materials, where the residual stress levels are low and concentrated near the surface, X-ray and photoelastic methods yield more

reliable results. However, in polycrystalline media with anisotropic grains, or more than one phase, differential thermo-elastic effects could generate higher levels of internal residual stresses which may be more amenable to ultrasonic velocity evaluation.

#### Publications and Technical Presentations

The results of this investigation are in the final stages of interpretation and evaluation. Two papers for technical journals are planned for preparation in the immediate future. The first will present the data, interpretation, and ramifications of the single-crystal spinel phase of the study. And the second will be based on the polycrystalline results and their implications for residual stress evaluation using acoustic methods. In addition, we plan to present the results of this work in three presentations at national scientific meetings. The first is planned for the annual western meeting of the AGU in December of this year. That talk will cover the polycrystalline results in regard to mineral elastic property measurements on hot-pressed aggregates. Two other presentations are planned for the forthcoming ACS meeting (polycrystalline and residual stress results) and national AGU meeting (single-crystal higher-order elastic properties).

#### Degrees Awarded

The work and results of this project also have provided the basis for three graduate degrees. Eric G. Hilbert has received already (Spring, 1984) an M.S. degree for his thesis, Ultrasonic Measurements of the Elastic Properties of Single-Crystal Magnesium Aluminate Spinel,  $\text{MgAl}_2\text{O}_4$ . Some of the higher-order elastic constant work on the single-crystal spinel will also constitute part of Mr. Hilbert's Ph.D. thesis, which is scheduled for completion by Spring, 1986. The polycrystalline results and residual stress

assessment will comprise the Ph.D. thesis of Mr. Blain Olbert. The first draft of the manuscript has been completed, and Mr. Olbert plans to complete his thesis by December, 1985.

# References

1. H.J. McSkimin, J. Acoust. Soc. Am., 33, 12 (1961).
2. H.J. McSkimin, P. Andreatch, J. Acoust. Soc. Am., 34, 609 (1962).
3. Z.P. Chang, G.R. Barsch, J. Geophys. Res., 78, 2418 (1973).
4. D.G. Isaak, E.K. Graham, J. Geophys. Res., 81, 2483 (1976).
5. Z.P. Chang, G.R. Barsch, D.R. Miller, Phys. Status Solidi, 23, 577 (1967).
6. E.H. Bogardus, J. Appl. Phys., 36, 2504 (1965).
7. W. Voigt, Lehrbuch der Kristallphysik, Teubner, Leipzig (1928).
8. A. Reuss, Z. Angew. Math. Mech., 9, 49 (1929).
9. R. Hill, Proc. Phys. Soc. London, Sect. A., 65, 349 (1952).
10. Z. Hashin, S. Shtrikman, J. Franklin Inst., 271, 336 (1961).
11. Z. Hashin, S. Shtrikman, J. Mech. Phys. Solids, 10, 335 (1962).
12. Z. Hashin, S. Shtrikman, J. Mech. Phys. Solids, 10, 343 (1962).
13. J.P. Watt, G.F. Davies, R.J. O'Connell, Rev. Geophys. Space Phys., 14, 541 (1976).
14. Z. Hashin, S. Shtrikman, J. Mech. Phys. Solids, 11, 127 (1963).
15. R.E. Green, Ultrasonic Investigation of Mechanical Properties, Academic Press (1973).
16. R.N. Thurston, J. Acoust. Soc. Am., 37, 348 (1965).
17. D.S. Hughes, J.L. Kelly, Phys. Rev., 92, 1145 (1953).
18. R.S. Crossin, J.W. Lin, J. Geophys. Res., 76, 570 (1971).
19. D.W. Baker, N.L. Carter, in Flow and Fracture of Rocks, Geophys. Monogr. Ser., 16. Amer. Geophys. Un., pp. 157-166 (1972).
20. J.D. Eshelby, Proc. Roy. Soc. London, Ser. A, 241, 376 (1957).
21. T.T. Wu, Int. J. Solid Struct., 3, 1 (1966).

22. G. Kuster, N. Toksoz, Geophysics, 39, 587 (1974).
23. D.L. Anderson, B. Minster, D. Cole, J. Geophys. Res., 79, 4011 (1974).
24. J.F. Nye, Physical Properties of Crystals, Oxford University Press, New York (1957).
25. G. Simmons, H. Wang, Single-Crystal Elastic Constants and Calculated Aggregate Properties: A handbook, Second Edition, M.I.T. Press, Cambridge, Mass. (1971).
26. M.F. Lewis, J. Acoust. Soc. Am., 40, 728 (1966).
27. R.J. O'Connell, E.K. Graham, EOS, Trans. Am. Geophys. Un., 52, 359 (1971).
28. H.P. Liu, R.N. Schock, D.L. Anderson, Geophys. J. R. Astr. Soc., 42, 217 (1975).